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## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner  
 US Department of Commerce  
 United States Patent and Trademark  
 Office, PCT  
 2011 South Clark Place Room  
 CP2/5C24  
 Arlington, VA 22202  
 ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

<b>Date of mailing (day/month/year)</b> 15 May 2001 (15.05.01)	<b>Applicant's or agent's file reference</b> P9966
<b>International application No.</b> PCT/NO00/00297	<b>Priority date (day/month/year)</b> 13 September 1999 (13.09.99)
<b>International filing date (day/month/year)</b> 11 September 2000 (11.09.00)	
<b>Applicant</b> LETH-OLSEN, Kari-Anne et al	

1. The designated Office is hereby notified of its election made:



in the demand filed with the International Preliminary Examining Authority on:

05 April 2001 (05.04.01)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

<b>The International Bureau of WIPO</b> 34, chemin des Colombettes 1211 Geneva 20, Switzerland  Facsimile No.: (41-22) 740.14.35	<b>Authorized officer</b>  G. Bähr  Telephone No.: (41-22) 338.83.38
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# PATENT COOPERATION TREATY

## PCT

### INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>P9966</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/NO 00/00297</b>	International filing date ( <i>day/month/year</i> ) <b>11 Sept 2000</b>	(Earliest) Priority Date ( <i>day/month/year</i> ) <b>13 Sept 1999</b>
Applicant <b>Norsk Hydro ASA et al</b>		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of   3   sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

**1. Basis of the report**

a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (See Box II).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No. \_\_\_\_\_

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☒ None of the figures.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 00/00297

## A. CLASSIFICATION OF SUBJECT MATTER

IPC7: C08F 291/00, C08F 2/08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7: C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	EP 0995764 A1 (AGFA-GEVAERT AG), 26 April 2000 (26.04.00), page 2, line 1 - page 3, line 20; page 5, line 30 - page 7, line 12, comparative examples 1-10, tables 1 and 3, abstract --	1-10
X	WO 9740076 A1 (NORSK HYDRO ASA), 30 October 1997 (30.10.97), page 3, abstract, examples --	1-10
Y ✓	EP 0326383 A2 (MITA INDUSTRIAL CO. LTD.), 2 August 1989 (02.08.89), abstract, examples, claims <i>ECT6.</i> --	1-10

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

## \* Special categories of cited documents:

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier application or patent but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

19 December 2000

Date of mailing of the international search report

20 -12- 2000

Name and mailing address of the ISA/  
Swedish Patent Office  
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Monika Bohlin/Els  
Telephone No. +46 8 782 25 00

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 00/00297

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 9831714 A1 (NORSK HYDRO ASA), 23 July 1998 (23.07.98), claims 1-3, abstract, examples --	1-10
Y	EP 0448391 A2 (ROHM AND HAAS COMPANY), 5,147,937 25 Sept 1991 (25.09.91) --	1-10
Y	US 4912184 A (YUTAKA AKASAKI ET AL), 27 March 1990 (27.03.90), abstract, examples, claims	1-10
A	US 4694035 A (KIYOSHI KASAI ET AL), 15 Sept 1987 (15.09.87), abstract --	1-10
A	US 5061766 A (HIROSHI YAMASHITA ET AL), 29 October 1991 (29.10.91), abstract, examples, claims --	1-10
A	US 4091054 A (FUMIO SAITO ET AL), 23 May 1978 (23.05.78), abstract -- -----	5

## INTERNATIONAL SEARCH REPORT

Information on patent family members

04/12/00

International application No.

PCT/NO 00/00297

Patent document cited in search report				Publication date		Patent family member(s)		Publication date	
EP	0995764	A1	26/04/00	DE	19848897	A		27/04/00	
				JP	2000128903	A		09/05/00	
-----									
WO	9740076	A1	30/10/97	AU	2653897	A		12/11/97	
				CA	2252514	A		30/10/97	
				EP	0897397	A		24/02/99	
				JP	2000509085	T		18/07/00	
				NO	308414	B		11/09/00	
				NO	961625	A		08/12/97	
				PL	329611	A		29/03/99	
				SK	146898	A		11/06/99	
-----									
EP	0326383	A2	02/08/89	DE	68916048	D,T		22/09/94	
				JP	1193303	A		03/08/89	
				JP	1968240	C		18/09/95	
				JP	6094483	B		24/11/94	
				KR	9308451	B		04/09/93	
				US	4996265	A		26/02/91	
				JP	1217004	A		30/08/89	
				JP	1960941	C		10/08/95	
				JP	6094484	B		24/11/94	
-----									
WO	9831714	A1	23/07/98	AU	5886198	A		07/08/98	
				EP	0954538	A		10/11/99	
				NO	970247	A		21/07/98	
-----									
EP	0448391	A2	25/09/91	SE	0448391	T3			
				AT	138400	T		15/06/96	
				AU	648293	B		21/04/94	
				AU	7361491	A		03/10/91	
				BR	9101115	A		05/11/91	
				CA	2038502	A		23/09/91	
				CN	1038756	B		17/06/98	
				CN	1059913	A		01/04/92	
				CZ	280984	B		15/05/96	
				CZ	9300778	A		19/01/94	
				DE	69119633	D,T		23/01/97	
				FI	911386	A		23/09/91	
				HU	61781	A		01/03/93	
				HU	910973	D		00/00/00	
				IL	97610	A		26/05/95	
				JP	3055071	B		19/06/00	
				JP	5093075	A		16/04/93	
				KR	171604	B		30/03/99	
				MX	173510	B		10/03/94	
				NO	911076	A		23/09/91	
				NZ	237483	A		27/07/93	
				PL	289528	A		16/12/91	
				PT	97102	A		29/11/91	
				US	5147937	A		15/09/92	
				ZA	9102111	A		25/03/92	
-----									

**INTERNATIONAL SEARCH REPORT**  
Information on patent family members

04/12/00

International application No.

PCT/NO 00/00297

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US <u>4912184 A</u>	27/03/90	JP 1959684 C	10/08/95
		JP 6096604 B	30/11/94
		JP 63191805 A	09/08/88
		JP 1876624 C	07/10/94
		JP 6002765 B	12/01/94
		JP 63191806 A	09/08/88
US <u>4694035 A</u>	15/09/87	EP 0190886 A	13/08/86
		JP 6074285 B	21/09/94
		JP 62121701 A	03/06/87
		NO 860171 A	31/07/86
		JP 61283602 A	13/12/86
US <u>5061766 A</u>	29/10/91	DE 3480992 D	00/00/00
		DE 3524179 A,C	16/01/86
		DE 8524879 U	02/01/86
		EP 0192772 A,B	03/09/86
		GB 2161170 A,B	08/01/86
		GB 8517247 D	00/00/00
		IT 1183953 B	22/10/87
		IT 8553770 V	12/09/85
		JP 6017373 B	09/03/94
		JP 61019602 A	28/01/86
		US 4627342 A	09/12/86
		US 4885350 A	05/12/89
		WO 8601774 A	27/03/86
US <u>4091054 A</u>	23/05/78	US 4174425 A	13/11/79

# PCT

LILLEGRAVEN, Rita  
NORSK HYDRO ASA  
N-0240 Oslo  
NORVEGE

## NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing (day/month/year)	11.12.2001
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Applicant's or agent's file reference  
P9966

## IMPORTANT NOTIFICATION

International application No.  
PCT/NO00/00297

International filing date (day/month/year)  
11/09/2000

Priority date (day/month/year)  
13/09/1999

Applicant  
NORSK HYDRO ASA et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

#### 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEAV

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## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference P9966		FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/NO00/00297	International filing date (day/month/year) 11/09/2000	Priority date (day/month/year) 13/09/1999	
International Patent Classification (IPC) or national classification and IPC C08F291/00			
Applicant NORSK HYDRO ASA et al.			
<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 5 sheets, including this cover sheet.</p> <p><input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of 5 sheets.</p>			
<p>3. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"><li>I <input checked="" type="checkbox"/> Basis of the report</li><li>II <input type="checkbox"/> Priority</li><li>III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</li><li>IV <input type="checkbox"/> Lack of unity of invention</li><li>V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</li><li>VI <input checked="" type="checkbox"/> Certain documents cited</li><li>VII <input checked="" type="checkbox"/> Certain defects in the international application</li><li>VIII <input checked="" type="checkbox"/> Certain observations on the international application</li></ul>			
Date of submission of the demand 05/04/2001		Date of completion of this report 11.12.2001	
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465		Authorized officer Boletti, C Telephone No. +49 89 2399 8527 	

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/NO00/00297

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, pages:**

1-3,6-13	as originally filed		
4,5,5a	as received on	14/09/2001	with letter of 10/09/2001

**Claims, No.:**

1-10	as received on	14/09/2001	with letter of 10/09/2001
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**Drawings, sheets:**

1/3-3/3	as originally filed
---------	---------------------

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/NO00/00297

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes: Claims
	No: Claims 1,7
Inventive step (IS)	Yes: Claims 2-6,8-10
	No: Claims
Industrial applicability (IA)	Yes: Claims 1-10
	No: Claims

- 2. Citations and explanations**  
**see separate sheet**

**VI. Certain documents cited**

- 1. Certain published documents (Rule 70.10)**

and / or

- 2. Non-written disclosures (Rule 70.9)**

**see separate sheet**

**VII. Certain defects in the international application**

The following defects in the form or contents of the international application have been noted:  
**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/NO00/00297

claims are fully supported by the description, are made:  
**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/NO00/00297

**ITEM V**

Reference is made to the following documents:

D1: WO 97/40076 (abstract, claims, tests B-1 to B-8 and Fig. 1)

D3: US 4694035 (claims, col. 2, l. 60-66, col. 5, l. 1-7, ex. 5-8,10,11 and Fig. 2-4)

The procedure referred to in claim 1 as well as the spherical polymer particles of claim 7 are known from D1 and D3.

Therefore, the subject-matter of claims 1 and 7 is not novel under Art. 33(2) PCT.

The subject-matter of claims 2 to 6 and 8 to 10 is either not novel with respect to D1/D3 or would not appear to be inventive with regard to these teachings (Art. 33 (2) and (3) PCT).

**ITEM VI**

It is noted that Doc. EP 0995764 (publ.: 26.04.2000) does not constitute prior art within the meaning of Rule 64.1(b) PCT. Nevertheless, this document could be relevant when the present application will enter in the regional phase. Particular attention should be given to p. 2, l. 3-4, p. 3, l. 13-20, p. 3, l. 55 to p. 4, l. 4, p. 9, l. 16-30 and Table 3.

**ITEM VII**

The amendments filed with the letter dated 10.09.2001 introduce subject-matter which extends beyond the content of the application as filed, contrary to Article 34(2)(b) PCT. The amendments concerned are the following: *wherein all the monomer is added directly to the start particles* in claim 1.

**ITEM VIII**

The subject-matter of claims 1 to 4 and 7 to 9 is not fully supported as such by the description (Art. 6 PCT).

## replacement sheet 4

structure can be formed in which the fraction of micro pores is insignificant. Another characteristic feature of the procedure is that the size distribution of the finished particles is narrow and in some cases narrower than in the start particles.

In order to describe prior art techniques in this field, reference will be made to the following patent specifications: NO 142082, NO 143403, NO 149108, NO 170730, NO 961625 (WO 97/40076), US 4,091,054, EP 0326383, EP 448391, US 4,382,124, EP 0 903 579 A1 and US 5,130,343.

NO 142082 and NO 143403 concern the technique that has become known as the two-step swelling method. The main element of this technique is that a low-molecular compound with very low water solubility is introduced into the start particles in a first stage. This results in an enormous increase in the particles' swelling capacity and much new monomer in relation to the polymer in the start particles can be absorbed and converted into polymer in a second step. The swelling ratio can be increased from 1:1-1:5 to 1:20-1:1000 times. NO 149108 also describes a two-stage swelling process. However, the special element of this method is that more swellable oligomer or oligomer-polymer particles are produced in the first stage. In these particles it is the oligomer content which produces an increased swelling capacity instead of introducing a low-molecular compound with very low water solubility as in NO 142082 and NO 143403. In NO 170730, the disadvantages of these techniques are mentioned as being that they involve several stages to produce polymer particles over 10  $\mu\text{m}$  since the start particles have a size of 0.5  $\mu\text{m}$ .

NO 170730 concerns a process for producing particles in the size range 1 to 30  $\mu\text{m}$ . In this process, start particles which are relatively large are used so that the swelling ratio can be small. Moreover, cross-linked start particles are used to avoid problems with coagulation, fusion and deformation of finished particles. A common feature with the first three references is that a first process stage is used involving the introduction of an activation agent (low-molecular compound with very low water solubility) to facilitate the absorption of new monomer in the second process stage. NO 961625 also uses relatively large start particles produced by dispersion polymerisation. Here, the total

replacement sheet 5

swelling ratio is also large, but to obtain spherical particles can be obtained, the majority of the monomer must be introduced during polymerisation.

US 4,091,054 concerns a process which takes place without introducing an activation agent for swelling in a separate step. An initiator solution and a monomer solution are dosed continuously but separately to start particles with a defined size range in such a way that all the polymer is formed within the start particles. Since the monomer is polymerised continuously, the ratio between the monomer and the polymer is always low. This produces controlled but limited growth of the particles.

EP 0326383 discloses a seeded polymerisation wherein the start particles are dispersed in an organic solvent in which also the monomer is dissolved. The organic solvent is miscible with water. The process is characterised in that the solubility of the monomer in the solvent is reduced thereby forcing the monomer into the start particles. Different means of reducing the solubility of the monomer in the solvent are described. It can be done by increasing the water concentration, by reducing the temperature, by adding water via a semipermeable membrane or by reducing the solvent concentration by evaporation.

Obvious disadvantages of this process are the use of large amounts of organic solvent and the need for measures to absorb the monomer into the start particles. The use of organic solvent will also to a large extent limit the versatility of this process to produce porous particles.

EP 448391 discloses a seed polymerisation using iterative steps to increase the particle size to the desired final particle size. Monomer is dosed in a strict controlled manner so the amount of free monomer in the system always is less than 10% of the amount of polymer present at any time during the polymerisation reaction. It is stated that if the amount of free monomer exceeds 10% there will be coagulation problems or there may be new formation of small particles which is highly undesirable. This gives a strong limitation of the process and it will never be possible to swell the polymer more than 1 time its own volume with new monomer. The total increase in diameter that is possible

## replacement sheet 5a

is 10 times. When, in addition, the start particles always are small (less than 2 microns are disclosed) a lot of iterative steps are necessary to achieve particles in the size range 10 to 50 microns.

US 4,382,124 concerns a traditional suspension polymerisation for the production of macroporous polymer particles. It describes how permanent porosity can be introduced into polymer particles. However, no measures are taken to control the size distribution or to prevent the formation of micro pores.

EP 0 903 579 A1 uses a technique very similar to that in NO 142082, NO 143403, NO 149108 and NO 170730 with regard to controlling the size and distribution of the particles but also describes new methods for avoiding micro pores. This is achieved by introducing, together with the monomer, a compound with conjugated double bonds which does not react with the monomers, or the use of an oxidation-reduction reaction, or heat treatment of the finished particles.

US 5,130,343 also concerns a process similar to that in NO 142082, NO 143403, NO 149108 and NO 170730 with regard to controlling the particle size and distribution. A soluble polymer, which is used as the start particle, is extracted from the finished particle to form a macroporous structure.

In accordance with the present invention, unusual and unexpected results have surprisingly been found when a seed polymerisation is performed in which the start particles are produced by dispersion polymerisation. It is particularly



**AMENDED CLAIMS**

1. A procedure for producing spherical polymer particles with a narrow size distribution, i.e. with a CV of less than 35%, preferably less than 20%, in the range between 5 and 100  $\mu\text{m}$  by free radical polymerisation of vinyl monomers in water, characterised in that the polymerisation is performed as a one-step seed polymerisation wherein all of the monomer is added directly to the start particles which consist of a non-cross-linked polymer produced by a dispersion polymerisation which gives the start particles such a high swelling capacity that they can absorb more than 5 times and preferably more than 20 times their own volume.
2. A procedure in accordance with claim 1, characterised in that the start particles absorb from 5 to 120, preferably from 20 to 100, times their own volume of vinyl monomers or a mixture which contains vinyl monomers.
3. A procedure in accordance with claim 2, characterised in that the mixture which contains vinyl monomers contains one or more inert solvents which lead to the formation of porosity in the polymer particles.
4. A procedure in accordance with claim 3, characterised in that the mixture which contains vinyl monomers also contains a polymerisation initiator in addition to one or more inert solvents.
5. A procedure in accordance with claim 4, characterised in that the polymerisation initiator is added separately from the mixture which contains vinyl monomers.
6. A procedure in accordance with claims 1 to 5, characterised in that the vinyl monomers or a mixture which contains vinyl monomers is finely divided into small emulsion droplets before they are swelled into the start particles.

# PCT

LILLEGRAVEN, Rita  
NORSK HYDRO ASA  
N-0240 Oslo  
NORVEGE

(PCT Rule 71.1)

## IMPORTANT NOTIFICATION

Priority date (day/month/year)  
13/09/1999

Applicant  
NORSK HYDRO ASA et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

#### 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Authorized officer

Le Bolloch, C  
Tel. +49 89 2399-8091



7. Spherical polymer particles with a narrow size distribution, i.e. with a CV of less than 35%, preferably less than 20%, in the range between 5 and 100  $\mu\text{m}$ , characterised in that they are produced by the procedure in accordance with claims 1-6.
8. Polymer particles in accordance with claim 7, characterised in that the porous structure in the particles has a very low content of pores with a diameter below 50 Å, i.e. less than 10%, preferably less than 5%, of the total pore volume.
9. Polymer particles in accordance with claim 7 or 8, characterised in that the porous structure in the particles is free from micropores with a diameter below 5 Å.
10. Polymer particles in accordance with claim 7, 8 or 9, characterised in that the size distribution of the polymer particles is always narrow and preferably narrower than that of the start particles.

PCT

28 SEPT 2000

For receiving Office use only

International Application No. PCT/NO 00 00297

(11.09.00) 11 SEPT. 2000

International Filing Date



PATENTET YRET

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference

(if desired) (12 characters maximum) P9966

## REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

## Box No. I TITLE OF INVENTION

Single stage seed polymerisation for the production of large polymer particles with a narrow size distribution

## Box No. II APPLICANT

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

NORSK HYDRO ASA  
N-0240 Oslo  
Norway

☐ This person is also inventor.

Telephone No.

+47 22 53 21 00

Facsimile No.

+47 22 53 27 25

Teleprinter No.

State (that is, country) of nationality:

NO

State (that is, country) of residence:

NO

This person is applicant  
for the purposes of:

☐ all designated  
States

☒ all designated States except  
the United States of America

☐ the United States  
of America only

☐ the States indicated in  
the Supplemental Box

## Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

LETH-OLSEN, Kari-Anne  
Frognerlia 23  
N-3715 Skien  
Norway

This person is:

☐ applicant only

☒ applicant and inventor

☐ inventor only (If this check-box  
is marked, do not fill in below.)

State (that is, country) of nationality:

NO

State (that is, country) of residence:

NO

This person is applicant  
for the purposes of:

☐ all designated  
States

☐ all designated States except  
the United States of America

☒ the United States  
of America only

☐ the States indicated in  
the Supplemental Box

☒ Further applicants and/or (further) inventors are indicated on a continuation sheet.

## Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf  
of the applicant(s) before the competent International Authorities as:

☒ agent

☐ common representative

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

LILLEGRAVEN, Rita  
NORSK HYDRO ASA  
N-0240 Oslo  
Norway

Telephone No.

+47 22 91 04 84

Facsimile No.

+47 22 91 05 02

Teleprinter No.

☐ Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

KOP1 til orientering

## Continuation of Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

*If none of the following sub-boxes is used, this sheet should not be included in the request.*

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

PAULSEN, Øystein  
Furulund Terrasse 11  
N-3950 Brevik  
Norway

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:  
NO

State (that is, country) of residence:  
NO

This person is applicant for the purposes of:

- ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

PEDERSEN, Steinar  
Mindåstunet 7  
N-3712 Skien  
Norway

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:  
NO

State (that is, country) of residence:  
NO

This person is applicant for the purposes of:

- ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

SÆTHRE, Bård  
Orionveien 90  
N-3942 Porsgrunn  
Norway

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:  
NO

State (that is, country) of residence:  
NO

This person is applicant for the purposes of:

- ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

LARSEN, Rolf Olaf  
Nustadringen 15  
N-3970 Langesund  
Norway

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

State (that is, country) of nationality:  
NO

State (that is, country) of residence:  
NO

This person is applicant for the purposes of:

- ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

☐ Further applicants and/or (further) inventors are indicated on another continuation sheet.

## Box No.V DESIGNATION OF STATES

The following designations are hereby made under Rule 4.9(a) (mark the applicable check-boxes, at least one must be marked):

## Regional Patent

- ☒ **AP ARIPO Patent:** GH Ghana, GM Gambia, KE Kenya, LS Lesotho, MW Malawi, SD Sudan, SL Sierra Leone, SZ Swaziland, TZ United Republic of Tanzania, UG Uganda, ZW Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT
- ☒ **EA Eurasian Patent:** AM Armenia, AZ Azerbaijan, BY Belarus, KG Kyrgyzstan, KZ Kazakhstan, MD Republic of Moldova, RU Russian Federation, TJ Tajikistan, TM Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT
- ☒ **EP European Patent:** AT Austria, BE Belgium, CH and LI Switzerland and Liechtenstein, CY Cyprus, DE Germany, DK Denmark, ES Spain, FI Finland, FR France, GB United Kingdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, MC Monaco, NL Netherlands, PT Portugal, SE Sweden, and any other State which is a Contracting State of the European Patent Convention and of the PCT
- ☒ **OA OAPI Patent:** BF Burkina Faso, BJ Benin, CF Central African Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, GA Gabon, GN Guinea, GW Guinea-Bissau, ML Mali, MR Mauritania, NE Niger, SN Senegal, TD Chad, TG Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT (if other kind of protection or treatment desired, specify on dotted line)

## National Patent (if other kind of protection or treatment desired, specify on dotted line):

- |   |  |
|---|--|
| <input checked="" type="checkbox"/> <b>AE</b> United Arab Emirates                  | <input checked="" type="checkbox"/> <b>LR</b> Liberia  |
| <input checked="" type="checkbox"/> <b>AL</b> Albania                               | <input checked="" type="checkbox"/> <b>LS</b> Lesotho  |
| <input checked="" type="checkbox"/> <b>AM</b> Armenia                               | <input checked="" type="checkbox"/> <b>LT</b> Lithuania  |
| <input checked="" type="checkbox"/> <b>AT</b> Austria                               | <input checked="" type="checkbox"/> <b>LU</b> Luxembourg   |
| <input checked="" type="checkbox"/> <b>AU</b> Australia                             | <input checked="" type="checkbox"/> <b>LV</b> Latvia   |
| <input checked="" type="checkbox"/> <b>AZ</b> Azerbaijan                            | <input checked="" type="checkbox"/> <b>MA</b> Morocco  |
| <input checked="" type="checkbox"/> <b>BA</b> Bosnia and Herzegovina                | <input checked="" type="checkbox"/> <b>MD</b> Republic of Moldova  |
| <input checked="" type="checkbox"/> <b>BB</b> Barbados                              | <input checked="" type="checkbox"/> <b>MG</b> Madagascar   |
| <input checked="" type="checkbox"/> <b>BG</b> Bulgaria                              | <input checked="" type="checkbox"/> <b>MK</b> The former Yugoslav Republic of Macedonia                      |
| <input checked="" type="checkbox"/> <b>BR</b> Brazil                                | <input checked="" type="checkbox"/> <b>MN</b> Mongolia   |
| <input checked="" type="checkbox"/> <b>BY</b> Belarus                               | <input checked="" type="checkbox"/> <b>MW</b> Malawi   |
| <input checked="" type="checkbox"/> <b>CA</b> Canada                                | <input checked="" type="checkbox"/> <b>MX</b> Mexico   |
| <input checked="" type="checkbox"/> <b>CH and LI</b> Switzerland and Liechtenstein  | <input checked="" type="checkbox"/> <b>NO</b> Norway   |
| <input checked="" type="checkbox"/> <b>CN</b> China                                 | <input checked="" type="checkbox"/> <b>NZ</b> New Zealand  |
| <input checked="" type="checkbox"/> <b>CR</b> Costa Rica                            | <input checked="" type="checkbox"/> <b>PL</b> Poland   |
| <input checked="" type="checkbox"/> <b>CU</b> Cuba                                  | <input checked="" type="checkbox"/> <b>PT</b> Portugal   |
| <input checked="" type="checkbox"/> <b>CZ</b> Czech Republic                        | <input checked="" type="checkbox"/> <b>RO</b> Romania  |
| <input checked="" type="checkbox"/> <b>DE</b> Germany                               | <input checked="" type="checkbox"/> <b>RU</b> Russian Federation   |
| <input checked="" type="checkbox"/> <b>DK</b> Denmark                               | <input checked="" type="checkbox"/> <b>SD</b> Sudan  |
| <input checked="" type="checkbox"/> <b>DM</b> Dominica                              | <input checked="" type="checkbox"/> <b>SE</b> Sweden   |
| <input checked="" type="checkbox"/> <b>EE</b> Estonia                               | <input checked="" type="checkbox"/> <b>SG</b> Singapore  |
| <input checked="" type="checkbox"/> <b>ES</b> Spain                                 | <input checked="" type="checkbox"/> <b>SI</b> Slovenia   |
| <input checked="" type="checkbox"/> <b>FI</b> Finland                               | <input checked="" type="checkbox"/> <b>SK</b> Slovakia   |
| <input checked="" type="checkbox"/> <b>GB</b> United Kingdom                        | <input checked="" type="checkbox"/> <b>SL</b> Sierra Leone   |
| <input checked="" type="checkbox"/> <b>GD</b> Grenada                               | <input checked="" type="checkbox"/> <b>TJ</b> Tajikistan   |
| <input checked="" type="checkbox"/> <b>GE</b> Georgia                               | <input checked="" type="checkbox"/> <b>TM</b> Turkmenistan   |
| <input checked="" type="checkbox"/> <b>GH</b> Ghana                                 | <input checked="" type="checkbox"/> <b>TR</b> Turkey   |
| <input checked="" type="checkbox"/> <b>GM</b> Gambia                                | <input checked="" type="checkbox"/> <b>TT</b> Trinidad and Tobago  |
| <input checked="" type="checkbox"/> <b>HR</b> Croatia                               | <input checked="" type="checkbox"/> <b>TZ</b> United Republic of Tanzania                                    |
| <input checked="" type="checkbox"/> <b>HU</b> Hungary                               | <input checked="" type="checkbox"/> <b>UA</b> Ukraine  |
| <input checked="" type="checkbox"/> <b>ID</b> Indonesia                             | <input checked="" type="checkbox"/> <b>UG</b> Uganda   |
| <input checked="" type="checkbox"/> <b>IL</b> Israel                                | <input checked="" type="checkbox"/> <b>US</b> United States of America                                       |
| <input checked="" type="checkbox"/> <b>IN</b> India                                 |  |
| <input checked="" type="checkbox"/> <b>IS</b> Iceland                               |  |
| <input checked="" type="checkbox"/> <b>JP</b> Japan                                 | <input checked="" type="checkbox"/> <b>UZ</b> Uzbekistan   |
| <input checked="" type="checkbox"/> <b>KE</b> Kenya                                 | <input checked="" type="checkbox"/> <b>VN</b> Viet Nam   |
| <input checked="" type="checkbox"/> <b>KG</b> Kyrgyzstan                            | <input checked="" type="checkbox"/> <b>YU</b> Yugoslavia   |
| <input checked="" type="checkbox"/> <b>KP</b> Democratic People's Republic of Korea | <input checked="" type="checkbox"/> <b>ZA</b> South Africa   |
|   | <input checked="" type="checkbox"/> <b>ZW</b> Zimbabwe   |
| <input checked="" type="checkbox"/> <b>KR</b> Republic of Korea                     | Check-boxes reserved for designating States which have become party to the PCT after issuance of this sheet: |
| <input checked="" type="checkbox"/> <b>KZ</b> Kazakhstan                            | <input type="checkbox"/>   |
| <input checked="" type="checkbox"/> <b>LC</b> Saint Lucia                           | <input type="checkbox"/>   |
| <input checked="" type="checkbox"/> <b>LK</b> Sri Lanka                             |  |

**Precautionary Designation Statement:** In addition to the designations made above, the applicant also makes under Rule 4.9(b) all other designations which would be permitted under the PCT except any designation(s) indicated in the Supplemental Box as being excluded from the scope of this statement. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation (including fees) must reach the receiving Office within the 15-month time limit.)

<b>Box No. VI PRIORITY CLAIM</b>					<input type="checkbox"/> Further priority claim is indicated in the Supplemental Box.
Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is:			
		national application: country	regional application: regional Office	international application: receiving Office	
item (1) 13. September 1999 (13.09.1999)	19994425	NO			
item (2)					
item (3)					

☒ The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) (only if the earlier application was filed with the Office which for the purposes of the present international application is the receiving Office) identified above as item(s): 1

\* Where the earlier application is an ARIPO application, it is mandatory to indicate in the Supplemental Box at least one country party to the Paris Convention for the Protection of Industrial Property for which that earlier application was filed (Rule 4.10(b)(ii)). See Supplemental Box.

<b>Box No. VII INTERNATIONAL SEARCHING AUTHORITY</b>									
<b>Choice of International Searching Authority (ISA)</b> (if two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used):  ISA / SE	<b>Request to use results of earlier search; reference to that search (if an earlier search has been carried out by or requested from the International Searching Authority):</b>  <table style="width: 100%;"> <tr> <td style="width: 33%;">Date (day/month/year)</td> <td style="width: 33%;">Number</td> <td style="width: 33%;">Country (or regional Office)</td> </tr> <tr> <td>12.02.2000</td> <td>1999 4425</td> <td>NO</td> </tr> </table>			Date (day/month/year)	Number	Country (or regional Office)	12.02.2000	1999 4425	NO
Date (day/month/year)	Number	Country (or regional Office)							
12.02.2000	1999 4425	NO							

<b>Box No. VIII CHECK LIST; LANGUAGE OF FILING</b>	
This international application contains the following number of sheets: request : 4 description (excluding sequence listing part) : 13 claims : 2 abstract : 1 drawings : 3 sequence listing part of description : 0 Total number of sheets : 23	This international application is accompanied by the item(s) marked below: 1. <input checked="" type="checkbox"/> fee calculation sheet 2. <input checked="" type="checkbox"/> separate signed power of attorney 3. <input type="checkbox"/> copy of general power of attorney; reference number, if any: 4. <input type="checkbox"/> statement explaining lack of signature 5. <input type="checkbox"/> priority document(s) identified in Box No. VI as item(s): 6. <input type="checkbox"/> translation of international application into (language): 7. <input type="checkbox"/> separate indications concerning deposited microorganism or other biological material 8. <input type="checkbox"/> nucleotide and/or amino acid sequence listing in computer readable form 9. <input checked="" type="checkbox"/> other (specify): Norwegian Search Report
<b>Figure of the drawings which should accompany the abstract:</b>	<b>Language of filing of the international application:</b> English

<b>Box No. IX SIGNATURE OF APPLICANT OR AGENT</b>	
<small>Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).</small>	
Rita Lillegraven  	

<b>For receiving Office use only</b>		2. Drawings:  <input checked="" type="checkbox"/> received:  <input type="checkbox"/> not received:
1. Date of actual receipt of the purported international application:	11 SEPT. 2000 (11.09.00)	
3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:		
4. Date of timely receipt of the required corrections under PCT Article 11(2):		
5. International Searching Authority (if two or more are competent): ISA/SE	6. <input type="checkbox"/> Transmittal of search copy delayed until search fee is paid.	

<b>For International Bureau use only</b>
Date of receipt of the record copy by the International Bureau:

## PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION CONCERNING  
SUBMISSION OR TRANSMITTAL  
OF PRIORITY DOCUMENT

(PCT Administrative Instructions, Section 411)

To:

LILLEGRAVEN, Rita  
Norsk Hydro ASA  
N-0240 Oslo  
NORVÈGE

Date of mailing (day/month/year) 25 October 2000 (25.10.00)	<b>IMPORTANT NOTIFICATION</b>
Applicant's or agent's file reference P9966	
International application No. PCT/NO00/00297	International filing date (day/month/year) 11 September 2000 (11.09.00)
International publication date (day/month/year) Not yet published	Priority date (day/month/year) 13 September 1999 (13.09.99)
Applicant NORSK HYDRO ASA et al	

- The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).
- This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.
- An asterisk(\*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.
- The letters "NR" appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, as provided by Rule 17.1(a) or (b), respectively. In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

<u>Priority date</u>	<u>Priority application No.</u>	<u>Country or regional Office or PCT receiving Office</u>	<u>Date of receipt of priority document</u>
13 Sept 1999 (13.09.99)	19994425	NO	29 Sept 2000 (29.09.00)

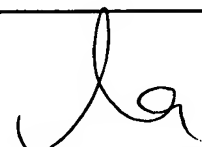
The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

N. Wagner

Telephone No. (41-22) 338.83.38





# PATENT COOPERATION TREATY

WO 01/19885  
PCT/NO00/0029

PCT

## NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

From the INTERNATIONAL BUREAU

To:

LILLEGRAVEN, Rita  
Norsk Hydro ASA  
N-0240 Oslo  
NORVÈGE

MOTTATT I N.H.  
PATENTAVD.  
30 MAR 2001

Date of mailing (day/month/year) 22 March 2001 (22.03.01)		IMPORTANT NOTICE	
Applicant's or agent's file reference P9966			
International application No. PCT/NO00/00297	International filing date (day/month/year) 11 September 2000 (11.09.00)	Priority date (day/month/year) 13 September 1999 (13.09.99)	
Applicant NORSK HYDRO ASA et al			

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:  
AU,KP,KR,US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:

AE,AL,AM,AP,AT,AZ,BA,BB,BG,BR,BY,CA,CH,CN,CR,CU,CZ,DE,DK,DM,EA,EE,EP,ES,FI,GB,GD,  
GE,GH,GM,HR,HU,ID,IL,IN,IS,JP,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MA,MD,MG,MK,MN,MW,MX,  
NO,NZ,OA,PL,PT,RO,RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,TZ,UA,UG,UZ,VN,YU,ZA,ZW

The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on  
22 March 2001 (22.03.01) under No. WO 01/19885

### REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a demand for international preliminary examination must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

### REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the national phase, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer  J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38

MOTTATT I N.H.  
PATENTAVD.

21 MAI 2001

# PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

To:

LILLEGRAVEN, Rita  
Norsk Hydro ASA  
N-0240 Oslo  
NORVÈGE

## INFORMATION CONCERNING ELECTED OFFICES NOTIFIED OF THEIR ELECTION

(PCT Rule 61.3)

Date of mailing (day/month/year) 15 May 2001 (15.05.01)		
Applicant's or agent's file reference P9966		IMPORTANT INFORMATION
International application No. PCT/NO00/00297	International filing date (day/month/year) 11 September 2000 (11.09.00)	
Priority date (day/month/year) 13 September 1999 (13.09.99)		
Applicant NORSK HYDRO ASA et al		

1. The applicant is hereby informed that the International Bureau has, according to Article 31(7), notified each of the following Offices of its election:

AP : GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW

EP : AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE

National : AU, BG, CA, CN, CZ, DE, IL, JP, KP, KR, MN, NO, NZ, PL, RO, RU, SE, SK, US

2. The following Offices have waived the requirement for the notification of their election; the notification will be sent to them by the International Bureau only upon their request:

EA : AM, AZ, BY, KG, KZ, MD, RU, TJ, TM

OA : BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG

National : AE, AL, AM, AT, AZ, BA, BB, BR, BY, CH, CR, CU, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IN, IS, KE, KG, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MW, MX, PT, SD, SG, SI, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW

3. The applicant is reminded that he must enter the "national phase" before the expiration of 30 months from the priority date before each of the Offices listed above. This must be done by paying the national fee(s) and furnishing, if prescribed, a translation of the international application (Article 39(1)(a)), as well as, where applicable, by furnishing a translation of any annexes of the international preliminary examination report (Article 36(3)(b) and Rule 74.1).

Some offices have fixed time limits expiring later than the above-mentioned time limit. For detailed information about the applicable time limits and the acts to be performed upon entry into the national phase before a particular Office, see Volume II of the PCT Applicant's Guide.

The entry into the European regional phase is postponed until 31 months from the priority date for all States designated for the purposes of obtaining a European patent.

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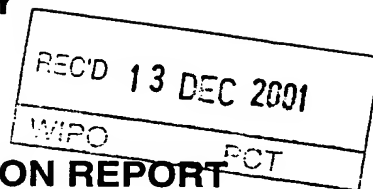
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# PATENT COOPERATION TREATY

## PCT

### INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)



14


Applicant's or agent's file reference <b>P9966</b>	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/NO00/00297</b>	International filing date (day/month/year) <b>11/09/2000</b>	Priority date (day/month/year) <b>13/09/1999</b>
International Patent Classification (IPC) or national classification and IPC <b>C08F291/00</b>		
Applicant <b>NORSK HYDRO ASA et al.</b>		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 5 sheets, including this cover sheet.
  - ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☒ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  <b>05/04/2001</b>	Date of completion of this report  <b>11.12.2001</b>
Name and mailing address of the international preliminary examining authority:   <b>European Patent Office</b> <b>D-80298 Munich</b> <b>Tel. +49 89 2399 - 0 Tx: 523656 epmu d</b> <b>Fax: +49 89 2399 - 4465</b>	Authorized officer  <b>Boletti, C</b>  Telephone No. <b>+49 89 2399 8527</b>



# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/NO00/00297

## I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

### Description, pages:

1-3,6-13	as originally filed		
4,5,5a	as received on	14/09/2001	with letter of 10/09/2001

### Claims, No.:

1-10	as received on	14/09/2001	with letter of 10/09/2001
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### Drawings, sheets:

1/3-3/3	as originally filed
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2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/NO00/00297

- ☐ the description,      pages:
- ☐ the claims,      Nos.:
- ☐ the drawings,      sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

## V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### 1. Statement

Novelty (N)	Yes:	Claims	
	No:	Claims	1,7
Inventive step (IS)	Yes:	Claims	2-6,8-10
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-10
	No:	Claims	

2. Citations and explanations  
**see separate sheet**

## VI. Certain documents cited

1. Certain published documents (Rule 70.10)

and / or

2. Non-written disclosures (Rule 70.9)

**see separate sheet**

## VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:  
**see separate sheet**

## VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/NO00/00297

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claims are fully supported by the description, are made:  
**see separate sheet**

## **ITEM V**

Reference is made to the following documents:

D1: WO 97/40076 (abstract, claims, tests B-1 to B-8 and Fig. 1)

D3: US 4694035 (claims, col. 2, l. 60-66, col. 5, l. 1-7, ex. 5-8,10,11 and Fig. 2-4)

The procedure referred to in claim 1 as well as the spherical polymer particles of claim 7 are known from D1 and D3.

Therefore, the subject-matter of claims 1 and 7 is not novel under Art. 33(2) PCT.

The subject-matter of claims 2 to 6 and 8 to 10 is either not novel with respect to D1/D3 or would not appear to be inventive with regard to these teachings (Art. 33 (2) and (3) PCT).

## **ITEM VI**

It is noted that Doc. EP 0995764 (publ.: 26.04.2000) does not constitute prior art within the meaning of Rule 64.1(b) PCT. Nevertheless, this document could be relevant when the present application will enter in the regional phase. Particular attention should be given to p. 2, l. 3-4, p. 3, l. 13-20, p. 3, l. 55 to p. 4, l. 4, p. 9, l. 16-30 and Table 3.

## **ITEM VII**

The amendments filed with the letter dated 10.09.2001 introduce subject-matter which extends beyond the content of the application as filed, contrary to Article 34(2)(b) PCT. The amendments concerned are the following: *wherein all the monomer is added directly to the start particles* in claim 1.

## **ITEM VIII**

The subject-matter of claims 1 to 4 and 7 to 9 is not fully supported as such by the description (Art. 6 PCT).

## replacement sheet 4

structure can be formed in which the fraction of micro pores is insignificant. Another characteristic feature of the procedure is that the size distribution of the finished particles is narrow and in some cases narrower than in the start particles.

In order to describe prior art techniques in this field, reference will be made to the following patent specifications: NO 142082, NO 143403, NO 149108, NO 170730, NO 961625 (WO 97/40076), US 4,091,054, EP 0326383, EP 448391, US 4,382,124, EP 0 903 579 A1 and US 5,130,343.

NO 142082 and NO 143403 concern the technique that has become known as the two-step swelling method. The main element of this technique is that a low-molecular compound with very low water solubility is introduced into the start particles in a first stage. This results in an enormous increase in the particles' swelling capacity and much new monomer in relation to the polymer in the start particles can be absorbed and converted into polymer in a second step. The swelling ratio can be increased from 1:1-1:5 to 1:20-1:1000 times. NO 149108 also describes a two-stage swelling process. However, the special element of this method is that more swellable oligomer or oligomer-polymer particles are produced in the first stage. In these particles it is the oligomer content which produces an increased swelling capacity instead of introducing a low-molecular compound with very low water solubility as in NO 142082 and NO 143403. In NO 170730, the disadvantages of these techniques are mentioned as being that they involve several stages to produce polymer particles over 10  $\mu\text{m}$  since the start particles have a size of 0.5  $\mu\text{m}$ .

NO 170730 concerns a process for producing particles in the size range 1 to 30  $\mu\text{m}$ . In this process, start particles which are relatively large are used so that the swelling ratio can be small. Moreover, cross-linked start particles are used to avoid problems with coagulation, fusion and deformation of finished particles. A common feature with the first three references is that a first process stage is used involving the introduction of an activation agent (low-molecular compound with very low water solubility) to facilitate the absorption of new monomer in the second process stage. NO 961625 also uses relatively large start particles produced by dispersion polymerisation. Here, the total



## replacement sheet 5

swelling ratio is also large, but to obtain spherical particles can be obtained, the majority of the monomer must be introduced during polymerisation.

US 4,091,054 concerns a process which takes place without introducing an activation agent for swelling in a separate step. An initiator solution and a monomer solution are dosed continuously but separately to start particles with a defined size range in such a way that all the polymer is formed within the start particles. Since the monomer is polymerised continuously, the ratio between the monomer and the polymer is always low. This produces controlled but limited growth of the particles.

EP 0326383 discloses a seeded polymerisation wherein the start particles are dispersed in an organic solvent in which also the monomer is dissolved. The organic solvent is miscible with water. The process is characterised in that the solubility of the monomer in the solvent is reduced thereby forcing the monomer into the start particles. Different means of reducing the solubility of the monomer in the solvent are described. It can be done by increasing the water concentration, by reducing the temperature, by adding water via a semipermeable membrane or by reducing the solvent concentration by evaporation.

Obvious disadvantages of this process are the use of large amounts of organic solvent and the need for measures to absorb the monomer into the start particles. The use of organic solvent will also to a large extent limit the versatility of this process to produce porous particles.

EP 448391 discloses a seed polymerisation using iterative steps to increase the particle size to the desired final particle size. Monomer is dosed in a strict controlled manner so the amount of free monomer in the system always is less than 10% of the amount of polymer present at any time during the polymerisation reaction. It is stated that if the amount of free monomer exceeds 10% there will be coagulation problems or there may be new formation of small particles which is highly undesirable. This gives a strong limitation of the process and it will never be possible to swell the polymer more than 1 time its own volume with new monomer. The total increase in diameter that is possible

# replacement sheet 5a

is 10 times. When, in addition, the start particles always are small (less than 2 microns are disclosed) a lot of iterative steps are necessary to achieve particles in the size range 10 to 50 microns.

US 4,382,124 concerns a traditional suspension polymerisation for the production of macroporous polymer particles. It describes how permanent porosity can be introduced into polymer particles. However, no measures are taken to control the size distribution or to prevent the formation of micro pores.

EP 0 903 579 A1 uses a technique very similar to that in NO 142082, NO 143403, NO 149108 and NO 170730 with regard to controlling the size and distribution of the particles but also describes new methods for avoiding micro pores. This is achieved by introducing, together with the monomer, a compound with conjugated double bonds which does not react with the monomers, or the use of an oxidation-reduction reaction, or heat treatment of the finished particles.

US 5,130,343 also concerns a process similar to that in NO 142082, NO 143403, NO 149108 and NO 170730 with regard to controlling the particle size and distribution. A soluble polymer, which is used as the start particle, is extracted from the finished particle to form a macroporous structure.

In accordance with the present invention, unusual and unexpected results have surprisingly been found when a seed polymerisation is performed in which the start particles are produced by dispersion polymerisation. It is particularly

## AMENDED CLAIMS

1. A procedure for producing spherical polymer particles with a narrow size distribution, i.e. with a CV of less than 35%, preferably less than 20%, in the range between 5 and 100  $\mu\text{m}$  by free radical polymerisation of vinyl monomers in water, characterised in that the polymerisation is performed as a one-step seed polymerisation wherein all of the monomer is added directly to the start particles which consist of a non-cross-linked polymer produced by a dispersion polymerisation which gives the start particles such a high swelling capacity that they can absorb more than 5 times and preferably more than 20 times their own volume.
2. A procedure in accordance with claim 1, characterised in that the start particles absorb from 5 to 120, preferably from 20 to 100, times their own volume of vinyl monomers or a mixture which contains vinyl monomers.
3. A procedure in accordance with claim 2, characterised in that the mixture which contains vinyl monomers contains one or more inert solvents which lead to the formation of porosity in the polymer particles.
4. A procedure in accordance with claim 3, characterised in that the mixture which contains vinyl monomers also contains a polymerisation initiator in addition to one or more inert solvents.
5. A procedure in accordance with claim 4, characterised in that the polymerisation initiator is added separately from the mixture which contains vinyl monomers.
6. A procedure in accordance with claims 1 to 5, characterised in that the vinyl monomers or a mixture which contains vinyl monomers is finely divided into small emulsion droplets before they are swelled into the start particles.

7. Spherical polymer particles with a narrow size distribution, i.e. with a CV of less than 35%, preferably less than 20%, in the range between 5 and 100  $\mu\text{m}$ , characterised in that they are produced by the procedure in accordance with claims 1-6.

8. Polymer particles in accordance with claim 7, characterised in that the porous structure in the particles has a very low content of pores with a diameter below 50 Å, i.e. less than 10%, preferably less than 5%, of the total pore volume.

9. Polymer particles in accordance with claim 7 or 8, characterised in that the porous structure in the particles is free from micropores with a diameter below 5 Å.

10. Polymer particles in accordance with claim 7, 8 or 9, characterised in that the size distribution of the polymer particles is always narrow and preferably narrower than that of the start particles.

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22 March 2001 (22.03.2001)

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**WO 01/19885 A1**

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- (71) Applicant (*for all designated States except US*): NORSK HYDRO ASA [NO/NO]; N-0240 Oslo (NO).
- (72) Inventors; and
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- For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

(54) Title: SINGLE STAGE SEED POLYMERISATION FOR THE PRODUCTION OF LARGE POLYMER PARTICLES WITH A NARROW SIZE DISTRIBUTION

(57) Abstract: The present invention concerns large spherical polymer particles with a narrow size distribution and a procedure for producing such particles. The procedure is characterised in that a seed polymerisation is performed in which the start particles have a very high ability to absorb new monomer so that finished particles are obtained after only one stage of polymerisation even if the finished particles are to be as large as in the range 10 to 100 µm. The procedure is also characterised in that, when porous particles are produced, a porous structure can be formed in which the fraction of micropores is insignificant. Another characteristic feature of the procedure is that the size distribution of the finished particles is narrow and in some cases narrower than in the start particles.

WO 01/19885 A1

Single stage seed polymerisation for the production of large polymer particles with a narrow size distribution

The present invention concerns large spherical polymer particles with a narrow  
5 size distribution and a procedure for producing such particles.

Polymer particles have today many areas of application such as separation, chromatography, adsorbents, ion exchangers, drug-delivery systems, solid-phase peptide synthesis, diagnostics and cosmetic preparations. Each application  
10 requires customised properties in the particles. The challenges involve controlling the particle size, the size distribution, the polymer composition, the porosity and the functionality.

Known techniques for the production of polymer particles are suspension  
15 polymerisation, emulsion polymerisation, miniemulsion polymerisation, microsuspension polymerisation and dispersion polymerisation. Suspension polymerisation is suitable for making large particles in the size range 0.05 to over 1 mm. The particle size is controlled by agitation and the type and concentration of suspending agents. The technique produces a very wide size distribution, which  
20 often limits the application possibilities. It is also difficult to achieve a desired size distribution when particles smaller than 100  $\mu\text{m}$  are to be produced. With miniemulsion and microsuspension polymerisation, prestabilised emulsion droplets of monomer are used to control the particle size. This also gives a very broad size distribution. With emulsion polymerisation, the particle size is controlled by the  
25 quantity and type of surfactants and not by mechanical agitation. This makes it possible to make very narrow size distributions, but the typical maximum particle size is approximately 1  $\mu\text{m}$ , and it is virtually impossible to make particles over approximately 10  $\mu\text{m}$  with this technique, which makes it unsuitable for making large polymer particles. Dispersion polymerisation differs from the above  
30 technologies in that the process is started in a homogeneous medium in which the monomer is soluble. When polymer is formed, it is precipitated as small particles that grow into the finished particles after the completion of the polymerisation. This process can produce narrow size distributions but in practice the maximum size is

limited to approximately 10  $\mu\text{m}$ . Attempts to produce larger particles have mostly resulted in broad size distributions. Another limitation in this method is the problem of obtaining particles with a high degree of cross-linking. In this system, the introduction of cross-linking produces a wider size distribution and a high probability for the particles to coagulate.

Seed polymerisation is a further development of the methods mentioned above. This technique starts with already polymerised particles, start particles. In principle, start particles can be produced by any method. The start particles are dispersed in the reaction medium and new monomer and initiator are added so that the original particles grow into larger particles in a controlled process. As shown above, it is not, however, easy to produce large polymer particles in the size range 10 to 100  $\mu\text{m}$ , or particles above 100  $\mu\text{m}$  with a narrow size distribution. Seed polymerisation is the most appropriate technique for performing this task. However, the method is very limited because the ability of the polymer particles to absorb new monomer is low. The general rule is that new monomer can be absorbed in the ratio 1:1, maximum 5:1, relative to the volume of the start particles.

The absorption of monomer can be described using a thermodynamic swelling equation (Morton equation). The driving force for the diffusion of the monomer into the start particles is the partial molar energy of the mixture of monomer and polymer. However, the particles can only absorb a limited amount of monomer before the equilibrium swelling is achieved. The reason for this is that the swelling leads to an increase in the surface area of the particles and thus to increased surface free energy. This effect, which thus counteracts the swelling, is inversely proportional to the particle radius. The equilibrium swelling of the monomer can be calculated from the expression:

$$\Delta G_m = RT[\ln(\phi)_m + (1 - 1/J_p)\phi_p + \phi_p^2\chi + 2V_m\gamma/rRT] = 0$$

where  $\Delta G_m$  is the total partial molar free energy of the mixture,  $\phi_m$  and  $\phi_p$  are the volume fractions of the monomer and polymer respectively,  $J_p$  is the polymer's chain length,  $V_m$  is the monomer's partial molar volume,  $r$  and  $\gamma$  are the particles' radius and interfacial tension and  $\chi$  is Flory-Huggin's interaction constant.

5

The development of polymer particles that have permanent porosity, macroporous particles, is particularly interesting for their use as adsorbents, ion exchangers and chromatography media. It is known that such particles can be obtained by using an inert solvent together with the monomer mixture to be polymerised. It is also

10 known that such solvents can be more or less good as solvents for the polymer formed. A soluble polymer can also be used as a pore forming agent. After the completion of polymerisation, the inert solvent and any soluble polymer are removed and a porous structure is obtained. In order to achieve a permanent porous structure, specific conditions must be present between the polymer formed  
15 and the solvent. The typical condition is that a cross-linked polymer is formed and that there is a complete phase separation between the polymer and the solvent. The size distribution of the pores formed is decisive for the area of application of the polymer particles. One known problem is that a considerable fraction of micropores less than 50 Å is formed. These are undesirable in separation  
20 processes because small molecules are left in the small pores and lead to poorer separation.

One main objective of the present invention is to produce compact and porous polymer particles and a procedure for producing such particles the above

25 disadvantages. This and other objectives of the present invention are achieved as described in the claims.

The procedure is characterised in that a seed polymerisation is performed in which the start particles have a very high ability to absorb new monomer so that finished

30 particles are obtained after only one polymerisation step even if the finished particles are to be as large as in the range 10 to 100 µm. The procedure is also characterised in that, when porous particles are produced, a porous structure can



be formed in which the fraction of micropores is insignificant. Another characteristic feature of the procedure is that the size distribution of the finished particles is narrow and in some cases narrower than in the start particles.

- 5 In order to describe prior art techniques in this field, reference will be made to the following patent specifications:

NO 142082, NO 143403, NO 149108, NO 170730, NO 961625, US 4,091,054, US 4,382,124, EP 0 903 579 A1 and US 5,130,343.

- 10 NO 142082 and NO 143403 concern the technique that has become known as the two-step swelling method. The main element of this technique is that a low-molecular compound with very low water solubility is introduced into the start particles in a first stage. This results in an enormous increase in the particles' swelling capacity and much new monomer in relation to the polymer in the start
- 15 particles can be absorbed and converted into polymer in a second step. The swelling ratio can be increased from 1:1-1:5 to 1:20-1:1000 times. NO 149108 also describes a two-stage swelling process. However, the special element of this method is that more swellable oligomer or oligomer-polymer particles are produced in the first stage. In these particles it is the oligomer content which
- 20 produces an increased swelling capacity instead of introducing a low-molecular compound with very low water solubility as in NO 142082 and NO 143403. In NO 170730, the disadvantages of these techniques are mentioned as being that they involve several stages to produce polymer particles over 10  $\mu\text{m}$  since the start particles have a size of 0.5  $\mu\text{m}$ .

25

- NO 170730 concerns a process for producing particles in the size range 1 to 30  $\mu\text{m}$ . In this process, start particles which are relatively large are used so that the swelling ratio can be small. Moreover, cross-linked start particles are used to avoid problems with coagulation, fusion and deformation of finished particles. A common
- 30 feature with the first three references is that a first process stage is used involving the introduction of an activation agent (low-molecular compound with very low water solubility) to facilitate the absorption of new monomer in the second process stage. NO 961625 also uses relatively large start particles produced by dispersion

polymerisation. Here, the total swelling ratio is also large, but to obtain spherical particles can be obtained, the majority of the monomer must be introduced during polymerisation.

- 5 US 4,091,054 concerns a process which takes place without introducing an activation agent for swelling in a separate step. An initiator solution and a monomer solution are dosed continuously but separately to start particles with a defined size range in such a way that all the polymer is formed within the start particles. Since the monomer is polymerised continuously, the ratio between the  
10 monomer and the polymer is always low. This produces controlled but limited growth of the particles.

- US 4,382,124 concerns a traditional suspension polymerisation for the production of macroporous polymer particles. It describes how permanent porosity can be  
15 introduced into polymer particles. However, no measures are taken to control the size distribution or to prevent the formation of micropores.

- EP 0 903 579 A1 uses a technique very similar to that in NO 142082, NO 143403, NO 149108 and NO 170730 with regard to controlling the size and distribution of  
20 the particles but also describes new methods for avoiding micropores. This is achieved by introducing, together with the monomer, a compound with conjugated double bonds which does not react with the monomers, or the use of an oxidation-reduction reaction, or heat treatment of the finished particles.

- 25 US 5,130,343 also concerns a process similar to that in NO 142082, NO 143403, NO 149108 and NO 170730 with regard to controlling the particle size and distribution. A soluble polymer, which is used as the start particle, is extracted from the finished particle to form a macroporous structure.

- 30 In accordance with the present invention, unusual and unexpected results have surprisingly been found when a seed polymerisation is performed in which the start particles are produced by dispersion polymerisation. It is particularly advantageous to use a dispersion polymerisation as described in NO 970247, which patent

specification is hereby included as a reference in the present invention. Detailed descriptions of dispersion polymerisation are also provided in K. E. Barret, Br. Polym. J., 5,259, 1973 and E. Shen et al., J of Pol. Sci., 32, 1087, 1994.

5 The basis for the present invention is an aqueous dispersion of start particles. To this dispersion is then added the monomer mixture which is to be polymerised to form the finished particles. If porous particles are to be produced, the monomer mixture contains one or more pore forming agents. The polymerisation initiator may also be part of the monomer mixture or it may be added separately before or  
10 after the addition of the monomer. The volume ratio between the monomer mixture and start particles is always greater than 5:1. This means that the start particles constitute maximum 20% of the product particles and that the increase in particle diameter from start particle to product particle is always greater than 1.8. When the monomer mixture and the initiator are absorbed in the start particles, the  
15 polymerisation is performed conventionally at a temperature suitable to the initiator. The product particles are also processed conventionally by dewatering and washing to remove the stabilisers used to prevent flocculation and coagulation during polymerisation, unconverted monomers and initiator and pore forming agents if such agents have been used to make porous particles.

20

The new, special feature of this procedure is that a large amount of monomer can be absorbed in the start particles without using any form of low-molecular compound with low water solubility as a swelling aid and without the swelling taking place in many stages or by continuous dosing of monomer. The new  
25 procedure is thus very simple and is performed in its entirety in just one cycle by the monomer being swelled into the start particles directly in the desired quantity and the polymerisation being performed. The quantity of monomer which can be swelled in may be varied from 5 to more than 100 times the volume of the start particles. From a start particle of, for example, 5  $\mu\text{m}$ , it is possible to produce  
30 products with diameters from approximately 9 to approximately 25  $\mu\text{m}$  and from a start particle of, for example, 15  $\mu\text{m}$ , it is possible to produce products with

diameters between 25 and 75  $\mu\text{m}$ . There are no restrictions in the procedure with regard to the type of polymerisation additives which may be used.

- The types and quantities of emulsifiers, polymer stabilisers, initiators, inhibitors, monomers and solvents can be chosen freely depending on the polymer composition, porosity and particle size to be achieved. Different methods for dosing the monomer mixture may also be chosen. Different monomer mixtures will have different transport velocities through the aqueous phase in which they are added and into the start particles. For monomer systems with low water solubility, it may be advantageous to add the monomer as finely divided droplets, as this increases the transport velocity into the start particles, or partially water-soluble compounds may be added which also increase the transport velocity of the monomer.
- Typical monomers used in the monomer mixture are aromatic and aliphatic vinyl monomers such as styrene, vinyl toluene, vinyl pyridine, methyl acrylate, methyl methacrylate, butyl acrylate, butyl methacrylate, phenyl acrylate, phenyl methacrylate, glycidyl methacrylate, hydroxy ethyl methacrylate, methacrylic acid, acrylic acid, acrylamide and cross-linking agents such as divinyl benzene, ethylene glycol dimethacrylate, diallyl phthalate, divinyl pyridine and trivinyl benzene.
- Emulsifiers and suspension agents are used to keep the particles freely dispersed during the process and possibly also to wet and disperse the start particles. Examples of suitable substances are sodium lauryl sulphate, ammonium laurate, ammonium myristate, sodium dodecyl benzene sulphonate, alkyl sulphonate, sodium lauryl ether sulphate, sodium dihexyl sulphosuccinate, sodium dioctyl sulphosuccinate, fatty alcohol ethoxylate, sorbitan esters, polyvinyl alcohol, polymer cellulose ethers, polyvinyl pyrrolidone, magnesium silicate and calcium phosphate. Common polymerisation initiators such as the peroxides: benzoyl peroxide, lauryl peroxide, tert-butyl perbenzoate, didecanoyl peroxide, dioctanoyl peroxide and cumene hydroperoxide and the azo-initiators: azodiisobutyronitrile, azobismethylbutyronitrile and azobisdimethylvaleronitrile can be used.

Another surprising feature of the procedure is that the product particles have been shown to be completely spherical and free from defects; nor are there any problems with deformation and fusion of particles during the seed polymerisation even though the start particles are a non-cross-linked polymer.

5

Yet another surprising feature of the procedure is that the size distribution of the product particles is always narrow and in some cases narrower than the distribution of the start particles. This produces special advantages and degrees of freedom with regard to the use of start particles with a relatively broad size

10 distribution even if a relatively narrow size distribution is required for the product.

This means that start particles produced by dispersion polymerisation which are above 10  $\mu\text{m}$  and have a relatively broad distribution, as discussed above, are also well suited for the production of particles above 50  $\mu\text{m}$  which are to have a narrow size distribution.

15

The fact that the size distribution is narrower when a monomer mixture is swelled into start particles produced by dispersion polymerisation with a relatively high diameter and a relatively wide size distribution conflicts with the theory of equilibrium swelling, described above, if it is assumed that the molecular weight

20 and surface conditions are equal for all start particles. The results indicate, therefore, that in a portion with start particles with a moderate size distribution, the smallest start particles will swell more than the largest start particles and the size will be equalised. This is very surprising.

25 Another surprising feature of the present invention is that the formation of micropores can be reduced considerably. This has not previously been possible without special measures, as described under the description of prior art techniques. One possible explanation is that the non-cross-linked polymer which constitutes the start particles contributes to a different phase separation in the  
30 system than when a cross-linked start particle is used.

The present invention will now be described in further detail using examples and figures that in no way restrict the possibilities of the present invention.

## DESCRIPTION OF FIGURES

Figure 1 shows a picture of the start particles from example A.1.

Figure 2 shows a picture of the start particles from example A.2.

5 Figure 3 shows a picture of the porous spherical polymer particles produced in accordance with example B.2.

Figure 4 shows a picture of the porous spherical polymer particles produced in accordance with example B.3.

10 Figure 5 shows a picture of the porous spherical polymer particles produced in accordance with example B.4.

The mean particle diameter and particle size distribution (CV) were determined using Coulter LS-230. CV is calculated by dividing the standard deviation (SD) by  
15 the mean particle diameter ( $d_p$ ):  $CV = (SD/d_p) * 100\%$ .

The specific surface area and pore volume were determined on the basis of  $N_2$  adsorption/desorption and a Hg porosimeter. A porosimeter (Units 120 and 2000) from Carlo Erba was used for the Hg measurements. The pore volume for pores  
20 with a diameter under 50 Å and pores with a diameter under 5 Å was determined on the basis of  $N_2$  adsorption/desorption using ASAP 2000 or Tristar 3000 from Micromeritics, USA.

## 25 EXAMPLES

### A. Production of start particles by dispersion polymerisation

Start particles were produced by dispersion polymerisation of methyl methacrylate  
30 in methanol with polyvinyl pyrrolidone (PVP K-30) as the stabiliser. Either 2,2-azobisisobutyronitrile (AIBN) or dioctanoyl peroxide was used as the initiator.

The following standard recipe was used:

Materials	Weight %
Methyl methacrylate	10 - 15
5 Methanol	80 - 85
PVP K-30	2.5 - 5
AIBN	0 - 0.4
Diocanoyl peroxide	0 - 0.4
Polymerisation temperature	50-58°C

10

Depending on the exact recipe, spherical particles were produced in the size range 1-15  $\mu\text{m}$  with a relatively narrow size distribution (CV 5-25%). When large start particles were produced, the size distribution was somewhat broader (CV 15-25%).

15

A constant polymerisation temperature was used throughout the polymerisation or the polymerisation temperature was controlled after the number of particles in the dispersion was constant (cf. patent NO 970247).

- 20 The following start particles were used for the production of large polymer particles with a narrow particle size distribution:

Example	Particle diameter ( $\mu\text{m}$ )	CV (%)
A.1	6	5
A.2	14	20
A.3	7	20

- Figures 1 and 2 show pictures of the particles from examples A.1 and A.2  
25 respectively.

B. One-step seed polymerisation for the production of large polymer particles with a narrow particle size distribution

Example B.1

5 A solution of methyl hydroxy propyl cellulose (0.7 g), sodium lauryl sulphate (0.5 g) and water (750 g) was mixed with a solution of styrene (38.0 g), ethylene glycol dimethacrylate (38.0 g), pentyl acetate (38 g) and azobismethylbutyronitrile (1.0 g). The mixture was emulsified using Ultra Turrax high-speed mixer and added to a reactor (5 l steel reactor). An aqueous dispersion of polymethyl methacrylate  
10 (PMMA) start particles produced in accordance with example A.1 (5.0 g start particles, 200 g water) and potassium iodide (0.2 g) were added to the reactor. The organic phase was left to swell into the start particles for 24 hours. Water (1500 g) was then added, after which the temperature was increased to 85°C. The polymerisation was completed in 7 hours at 85°C.

15

Porous spherical polymer particles with a mean diameter of 17  $\mu\text{m}$  and a CV of 16% were obtained. The specific surface area was 63  $\text{m}^2/\text{g}$  and the pore volume was 1.0 ml/g. The pore volume for pores with a diameter below 50 Å was 0.015 ml/g and for pores with a diameter less than 5 Å < 0.001 ml/g.

20

Example B.2

A solution of methyl hydroxy propyl cellulose (0.7 g), sodium lauryl sulphate (0.5 g) and water (750 g) was mixed with a solution of methyl methacrylate (28 g), ethylene glycol dimethacrylate (84 g), pentyl acetate (48 g) and  
25 azobismethylbutyronitrile (1.0 g). The mixture was emulsified using Ultra Turrax high-speed mixer and added to a reactor (5 l steel reactor). An aqueous dispersion of PMMA start particles produced in accordance with example A.1 (5 g start particles, 174 g water) and potassium iodide (0.2 g) were added to the reactor. The organic phase was left to swell into the start particles for 2 hours. Water (1500  
30 g) was then added, after which the temperature was increased to 80°C. The polymerisation was completed in 7 hours at 80°C.



Porous spherical polymer particles with a mean diameter of 18  $\mu\text{m}$  and a CV of 20% were obtained; see Figure 3. The specific surface area was 50  $\text{m}^2/\text{g}$  and the pore volume was 0.4  $\text{ml/g}$ . The pore volume for pores with a diameter below 50  $\text{\AA}$  was 0.011  $\text{ml/g}$  and for pores with a diameter less than 5  $\text{\AA}$  0.004  $\text{ml/g}$ .

5

### Example B.3

A solution of methyl hydroxy propyl cellulose (0.14 g), sodium lauryl sulphate (0.10 g) and water (110 g) was mixed with a solution of methyl methacrylate (7.60 g), ethylene glycol dimethacrylate (7.60 g), pentyl acetate (16.80 g) and  
10 azobismethylbutyronitrile (0.15 g). The mixture was emulsified using Ultra Turrax high-speed mixer and added to a reactor (0.5 l double-walled glass reactor). An aqueous dispersion of PMMA start particles produced in accordance with example A.2 (0.5 g start particles, 1.2 g water) was added to the reactor. The organic phase was left to swell into the start particles for 48 hours. Water (300 g) and potassium  
15 iodide (0.04 g) were then added, after which the temperature was increased to 80°C. The polymerisation was completed in 7 hours at 80°C.

Porous spherical polymer particles with a mean diameter of 55  $\mu\text{m}$  and a CV of 7% were obtained; see Figure 4. The specific surface area was 63  $\text{m}^2/\text{g}$  and the pore  
20 volume was 1.0  $\text{ml/g}$ . The pore volume for pores with a diameter below 50  $\text{\AA}$  was 0.011  $\text{ml/g}$  and for pores with a diameter less than 5  $\text{\AA}$  0.001  $\text{ml/g}$ .

### Example B.4

A solution of methyl hydroxy propyl cellulose (0.14 g), sodium lauryl sulphate (0.10  
25 g) and water (110 g) was mixed with a solution of styrene (7.60 g), ethylene glycol dimethacrylate (7.60 g), pentyl acetate (16.80 g) and azobismethylbutyronitrile (0.20 g). The mixture was emulsified using Ultra Turrax high-speed mixer and added to a reactor (0.5 l double-walled glass reactor). An aqueous dispersion of PMMA start particles produced in accordance with example A.2 (0.5 g start  
30 particles, 41.2 g water) was added to the reactor. The organic phase was left to swell into the start particles for 48 hours. Water (300 g) and potassium iodide (0.04 g) were then added, after which the temperature was increased to 80°C. The polymerisation was completed in 7 hours at 80°C.

Porous spherical polymer particles with a mean diameter of 50  $\mu\text{m}$  and a CV of 9% were obtained; see Figure 5. The specific surface area was 68  $\text{m}^2/\text{g}$  and the pore volume was 1.0  $\text{ml/g}$ . The pore volume for pores with a diameter below 50  $\text{\AA}$  was 0.017  $\text{ml/g}$  and for pores with a diameter less than 5  $\text{\AA}$  < 0.0001  $\text{ml/g}$ .

5

#### Example B.5

A solution of methyl hydroxy propyl cellulose (0.14 g), sodium lauryl sulphate (0.10 g) and water (110 g) was mixed with a solution of methyl methacrylate (4.8 g), ethylene glycol dimethacrylate (14.4 g), pentyl acetate (12.8 g) and  
10 azobismethylbutyronitrile (0.20 g). The mixture was emulsified using Ultra Turrax high-speed mixer and added to a reactor (0.5 l double-walled glass reactor). An aqueous dispersion of PMMA start particles produced in accordance with example A.3 (1.0 g start particles, 41.6 g water) and potassium iodide (0.04 g) were added to the reactor. The organic phase was left to swell into the start particles for 2  
15 hours. Water (300 g) was then added, after which the temperature was increased to 60°C. The polymerisation was completed in 7 hours at 60°C.

Porous spherical polymer particles with a mean diameter of 24  $\mu\text{m}$  and a CV of 18% were obtained. The specific surface area was 129  $\text{m}^2/\text{g}$  and the pore volume  
20 was 0.7  $\text{ml/g}$ . The pore volume for pores with a diameter below 50  $\text{\AA}$  was 0.045  $\text{ml/g}$  and for pores with a diameter less than 5  $\text{\AA}$  < 0.0001  $\text{ml/g}$ .

## CLAIMS

1. A procedure for producing spherical polymer particles with a narrow size distribution, i.e. with a CV of less than 35%, preferably less than 20%, in the range  
5 between 5 and 100  $\mu\text{m}$  by free radical polymerisation of vinyl monomers, characterised in that the polymerisation is performed as a one-step seed polymerisation in which the start particles consist of a non-cross-linked polymer produced by a dispersion polymerisation which gives the start particles such a high swelling capacity that they can absorb more than 5 times and  
10 preferably more than 20 times their own volume.
2. A procedure in accordance with claim 1, characterised in that the start particles absorb from 5 to 120, preferably from 20 to 100, times their own volume of vinyl monomers or a mixture which contains vinyl monomers.  
15
3. A procedure in accordance with claim 2, characterised in that the mixture which contains vinyl monomers contains one or more inert solvents which lead to the formation of porosity in the polymer particles.
- 20 4. A procedure in accordance with claim 3, characterised in that the mixture which contains vinyl monomers also contains a polymerisation initiator in addition to one or more inert solvents.
5. A procedure in accordance with claim 4, characterised in that  
25 the polymerisation initiator is added separately from the mixture which contains vinyl monomers.
6. A procedure in accordance with claims 1 to 5, characterised in that the vinyl monomers or a mixture which contains vinyl monomers is finely  
30 divided into small emulsion droplets before they are swelled into the start particles.

7. Spherical polymer particles with a narrow size distribution, i.e. with a CV of less than 35%, preferably less than 20%, in the range between 5 and 100  $\mu\text{m}$ , characterised in that they are produced by the procedure in accordance with claims 1-6.

5

8. Polymer particles in accordance with claim 7, characterised in that the porous structure in the particles has a very low content of pores with a diameter below 50  $\text{\AA}$ , i.e. less than 10%, preferably less than 5%, of the total pore volume.

10

9. Polymer particles in accordance with claim 7 or 8, characterised in that the porous structure in the particles is free from micropores with a diameter below 5  $\text{\AA}$ .

15 10. Polymer particles in accordance with claim 7, 8 or 9, characterised in that the size distribution of the polymer particles is always narrow and preferably narrower than that of the start particles.

1/3

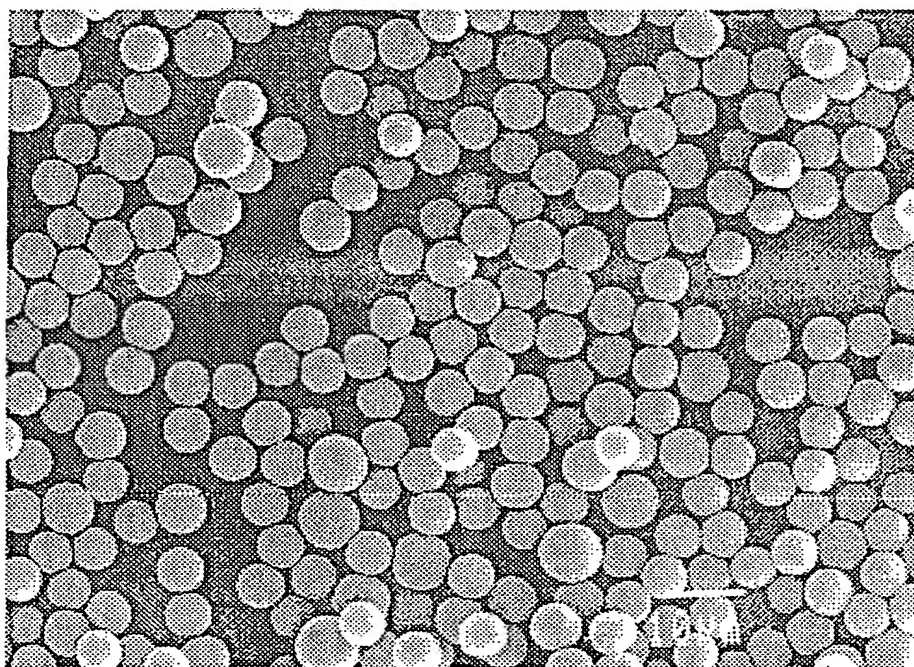


Fig. 1: SEM picture of start particles produced in accordance with example A.1

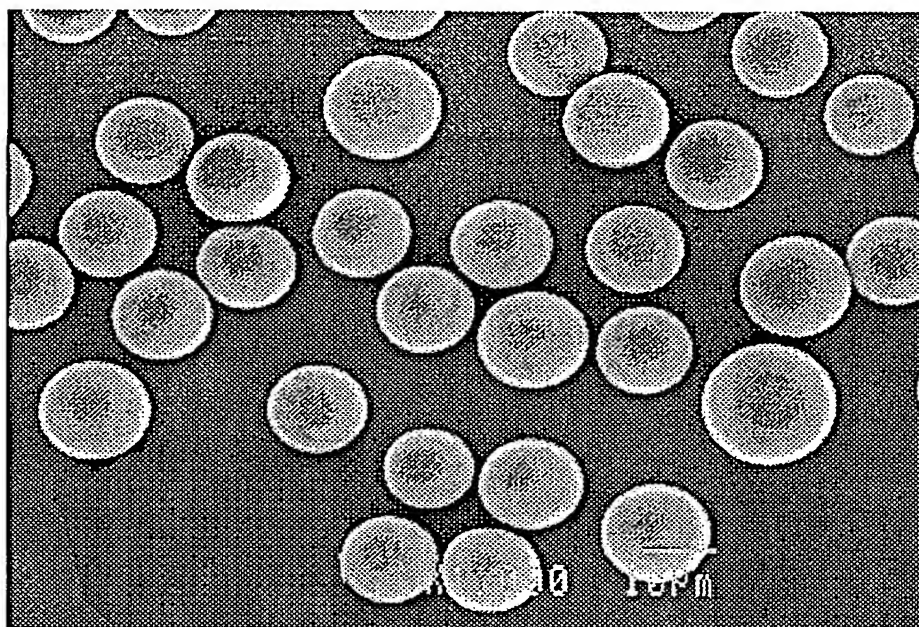


Fig.2: SEM picture of start particles produced in accordance with example A.2

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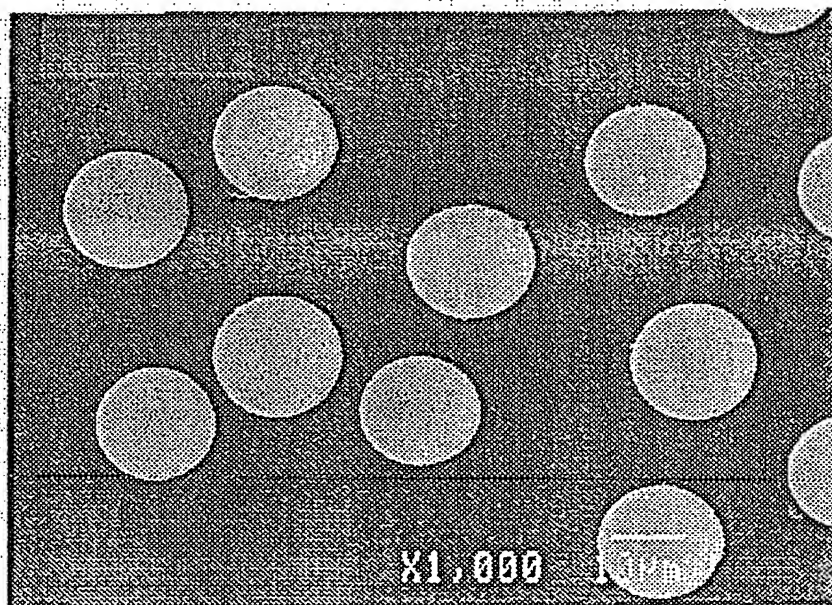


Fig. 3: SEM picture of polymer particles produced in accordance with example B.2

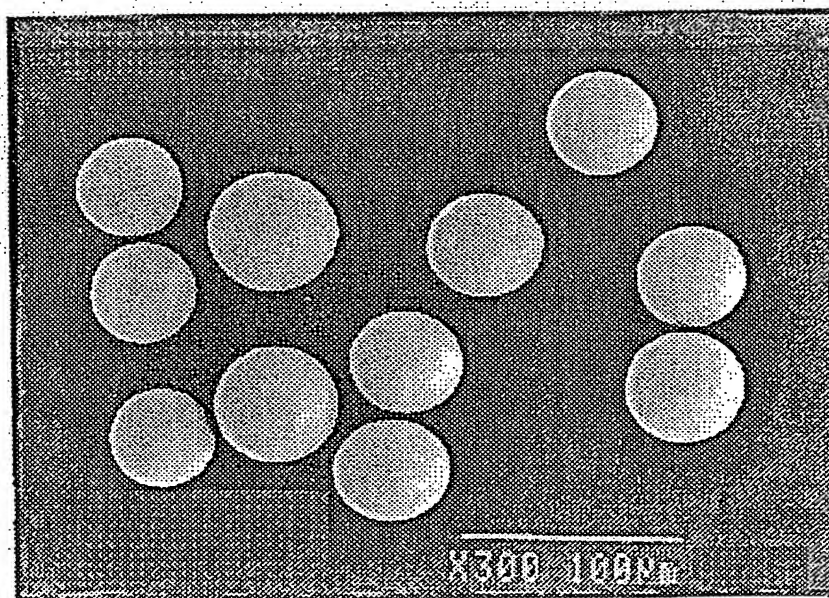


Fig. 4: SEM picture of porous particles produced in accordance with example B.3

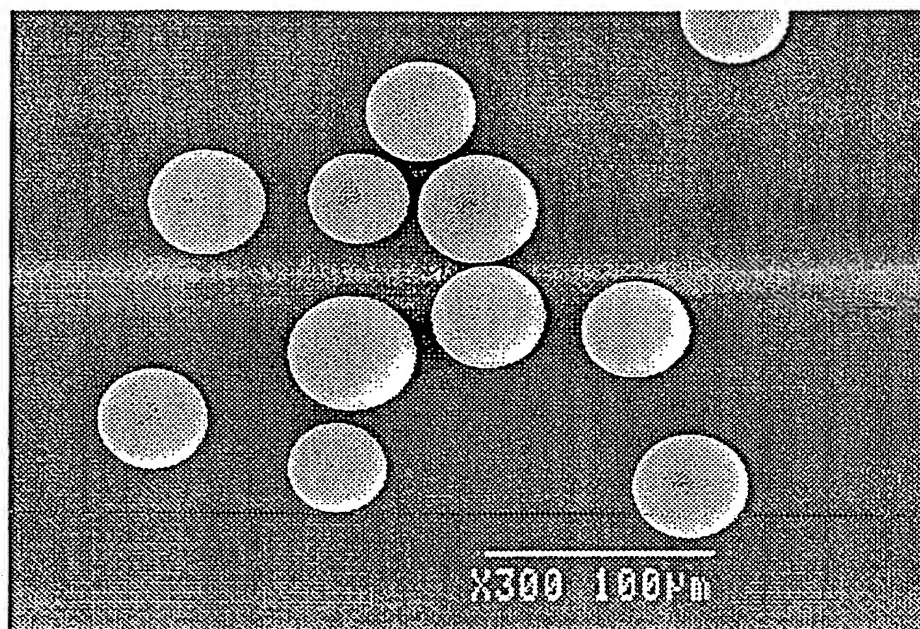


Fig. 5: SEM picture of porous particles produced in accordance with example B.4



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 00/00297

## A. CLASSIFICATION OF SUBJECT MATTER

IPC7: C08F 291/00, C08F 2/08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7: C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

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X	WO 9740076 A1 (NORSK HYDRO ASA), 30 October 1997 (30.10.97), page 3, abstract, examples --	1-10
Y	EP 0326383 A2 (MITA INDUSTRIAL CO. LTD.), 2 August 1989 (02.08.89), abstract, examples, claims --	1-10

☒ Further documents are listed in the continuation of Box C.
 ☒ See patent family annex.

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"&amp;" document member of the same patent family

Date of the actual completion of the international search

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## INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 00/00297

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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